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14. ABSTRACT

The properties of advanced materials and phenomena basic to the development of new and/or improved superconducting electronic technologies were studied. These include the conducting magnetic oxide strontium ruthenate relevant to high-temperature Josephson devices, the doped magnetic oxide lanthanum manganate relevant to magnetic sensing, and scanning tunneling microscopy of the superconducting energy gap in the high-temperature superconductors. We found that the transport properties of strontium ruthenate cannot be understood using the conventional theory of metals, that the spin polarized of 72% in tunnel junctions incorporating doped lanthanum manganate, and that there are intrinsic inhomogeneities in the superconducting energy gap of the high temperature superconductors on mesoscopic length scales.

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Final Technical Report

ADVANCED SUPERCONDUCTING MATERIALS AND DEVICE CONCEPTS

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July 2001

Final Technical Report

ADVANCED SUPERCONDUCTING MATERIALS AND DEVICE CONCEPTS

In this program we have studied the properties of a range of advanced materials and phenomena whose understanding are basic to the development of new and/or improved superconducting and other cryogenic electronic technologies. Particular emphasis was placed on materials and physics relevant to Josephson junction formation with the high temperature superconductors; the related issue of the role of defects on the superconducting energy gap in the high temperature superconductors; and magnetic materials and phenomena relevant to new device concepts for memory elements compatible with Josephson junction logic circuits.

Metallic oxides for cryogenic electronic applications.

$SrRuO_3$ is an itinerant ferromagnetic oxide. For reasons of materials compatibility, it is of interest both as a possible normal metal barrier in high temperature superconducting SNS junctions and as a ferromagnetic layer in a new superconducting memory element proposed by our group under other support. The theory underlying these two applications assumes conventional conduction mechanisms and conventional magnetism. In this program we have demonstrated that the magnetism in these materials can be understood using conventional theory, whereas the electrical conduction cannot. Hence our understanding of the physics of devices incorporating this material must be considered incomplete at best.

In our work we have established procedures to deposit thin films of $SrRuO_3$ both by electron beam thermal deposition and by pulsed laser deposition. The films made by e-beam deposition are outstanding and have resistance ratios exceeding even those of single crystals of this material. Transport measurements reveal that the resistivity of this material remains temperature dependent well above the temperature at which the electronic mean free path is less than one inter atomic distance. Such behavior is inconsistent with conventional (Boltzmann) transport theory. Similarly the frequency dependence of the conductivity of the material falls off as $f^{-1/2}$, as opposed to f^{-2}, as in conventional metals (Drude theory). On the other hand, the very low temperature magneto-resistance shows quantum oscillations in accord with a conventional metal with a well-defined Fermi surface. At present there is no theoretical understanding of the anomalous transport properties of this material.

We have also studied transport in the sister material $CaRuO_3$. Interestingly, this material is not ferromagnetic. However, it appears to show similarly anomalous transport properties.

Using electron beam lithography, we have patterned micron and sub-micron-wide thin film bridges of this material and studied the statistics of the threshold fields for magnetic switching. Switching was detected by its effect on the magneto-resistance and Hall effect voltage in the films. Evidence for thermal broadening of the distribution of switching fields has been observed. Further analysis will be carried out to look for quantum fluctuation switching at low temperatures, and the associated limits this distribution of switching fields puts on applications of the material in the devices mentioned above.

Magnetism and magnetic materials and devices.

$LaMnO_3$ doped with Ca or Sr exhibits a so-called colossal magnetoresistance that is of possible interest in magnetic memories and sensors. The conditions for depositing doped epitaxial $LaMnO_3$ thin films using pulsed laser deposition were established. Conductivity measurements from room temperature to 1200K showed that transport in $La_{1-x}Ca_xMnO_3$ over the entire doping range from x = 0 to 1 can be fit very well by the adiabatic small polaron theory of Emins and Holstein, in preference to the three other models in the literature. Further, the x-dependence of the prefactor of the conductivity show that the onsite coulomb repulsion plays an important role in the transport, while the asymmetry (as a function of x) shows that further refinements, including near-neighbor interactions, should also be included. The variation of the activation energy with x reflects the greater binding energy of the polaron by trivalent La than by divalent Ca.

The conditions for growing thin films of these materials is reproducible, and these films have been used to fabricate very well-behaved $La_{2/3}Sr_{1/3}MnO_3/SrTiO_3/Al$ tunneling junctions. Alternative tunnel barriers such as Al_20_3 and $Ce0_2$ gave less reproducible results than those obtained with $SrTiO_3$, presumably because pinholes (shorts) in the $SrTiO_3$ barriers are blocked by the much higher resistance of the Al_2O_3 which forms in the pinhole. Using these junctions and the approach of Tedrow and Meservey, the parameters of spin polarized tunneling resulting from the magnetism of the $LaMnO_3$ counter electrode were established. The parameters have important implications for the application of this material for in-magnetic sensors, in particular disc drive read heads.

The theory of Maki, including orbital depairing, spin-orbit scattering, and Zeeman splitting of the spin states in the Al base electrode, was used to fit the tunneling data and obtain a quantitative fit of the conductance. The reproducible value for the spin polarization in doped $LaMnO_3$ (defined as the difference between the up and down spin tunneling currents) is 72%.

Similar spin polarized tunneling experiments were carried out on ferromagnetic $SrRuO_3$ electrodes. In contrast to results in the literature on 12 ferromagnetic materials studied by the Tedrow-Meservey method, the spin polarized current from $SrRuO_3$ is dominated by minority as opposed to majority spins. The dependence of these inferred degrees of spin polarization depend on the material used in the tunneling barrier will require further investigation.

Inhomogeneous Gap Structure in HTSC

Tunneling spectroscopy has been an important tool in the study of high-temperature superconductors since their discovery. The magnitude, orbital symmetry and uniformity of the superconducting energy gap directly affect the performance and homogeneity of all Josephson junction devices. In the early days of the field, tunneling studies revealed a wide variety of tunneling I-V curves and inferred energy gaps. Much controversy surrounded these results. More recently better materials and tunneling approaches based on scanning tunneling microscopy (STM) probes have yielded greater consistency among groups, and a more coherent picture of the surface of the high-T_c superconductors. Among many examples we note that STM studies revealed the nature of the superstructure in BSCCO, the size and d-wave nature of the superconducting energy gap, the effect of local impurities, the emergence of zero-bias anomalies, and the electronic structure in the cores of vortices.

Despite all this work, very little has been published on the large-scale variations of the electronic state at the surface of high-T_c materials. Judging from photoemission measurements, which clearly reveal a **k**-dependent gap and therefore imply that k is a good quantum number, it has been the common belief that the surface is homogeneous and therefore represents the bulk behavior of the material. However, recent measurements by a variety of techniques suggest that superconductivity may not be homogeneous in high-T_c superconductors. In particular, STM measurements of low-temperature-cleaved YBCO have found that the gap along the chains is inhomogeneous and correlates with oxygen vacancies. In addition to practical device considerations, the issue of homogeneity (either static or dynamic) is very important in view of recent theoretical developments that find phase separation to be an integral part of the high-T_c scenario.

Our main recent result was scanning tunneling spectroscopy measurements on the surface of $Bi_2Sr_2CaCu_2O_{8-x}$ (BSCCO) single crystals which show that superconductivity in at least this particular high-temperature superconductor may be inherently inhomogeneous. This inhomogeneity appears in underdoped BSCCO at a scale of a few coherence lengths (10 to 30 Angstroms) and is not a consequence of local impurities or strong disorder. We have argued that this inhomogeneity is a consequence of an electronic phase separation into superconducting and insulating regions that are proximity-coupled to give a continuous variation of the tunneling spectroscopic features. In addition, the doping dependence shows that the effect persists into overdoped material, but with decreased inhomogeneity, despite the increase in disorder in such overdoped samples due to the disrupted superstructure.

A high-speed pattern recognition circuit using series arrays of Josephson junctions.

Finally, as part of this program, we conceived of a new approach to analog correlation (pattern recognition) using Josephson junction arrays. This is an entirely new circuit concept. Using computer simulations, we confirmed this circuit concept and further explored its characteristics in recognizing 32×32 bit 2D patterns with and without the influence of noise.

PERSONNEL SUPPORTED

Faculty—Professors M. R. Beasley, T. H. Geballe, and A. Kapitulnik
Sr. Research Scientist—Dr. R.H. Hammond
Visiting Researcher Associates: Drs. B. Czyzak, S. Kashiwaya, M. Barahona
Postdoctoral Researchers: L. Mieville, S. Döttinger
Students (primary): J. Chiaverini, A. Fang, C. Howald, N. Ingle, L. Litvak, J. Reiner, M. Rozler, D. Sisson, C. Wang, D. Worledge.
Students (partial): none
Rotation and Summer Students: Mollie Uhl, Alex Panchula D. Freedman, W. Zheng

PUBLICATIONS

Accepted:

1. D.C. Worledge, L. Mieville, and T.H. Geballe, "On-site Coulomb Repulsion in the Small Polaron System $La_cCa_{1-c}MnO_3$," *Phys. Rev. B* **57**, 15267-15271 (June 1998).

2. D.C. Worledge, L. Mieville, and T.H. Geballe, "Growth and Small Polaron Properties of Epitaxial $La_{1-x}Ca_xMnO_3$ Thin Films," *J. Appl. Phys.* **83**, 5913-5916 (June 1998).

3. L. Mieville, D.C. Worledge, T.H. Geballe, R. Contreras, and K. Char, "Transport across conducting ferromagnetic oxide/metal interfaces," *Phys. Rev. Lett.* (in press).

4. P. Kostic, Y. Okada, Z. Schlesinger, J.W. Reiner, L. Klein, A. Kapitulnik, T.H. Geballe, and M.R. Beasley, *Phys. Rev. Lett.*(in press).

5. A. Yazdani, C.M. Howald, C.P. Lutz, A. Kapitulnik and D.M. Eigler, *Phys. Rev. Lett.* **83**, 176 (1999).

6. L. Mieville, D.C. Worledge, T.H. Geballe, R. Contreras, and K. Char, "Transport across conducting ferromagnetic oxide/metal interfaces," *Appl. Phys. Lett.* **73**, 1736-1738 (Sept. 21, 1998).

7. A.P. Mackenzie, J.W. Reiner, A.W. Tyler, L.M. Galvin, S.R. Julian, M.R. Beasley, T.H. Geballe, and A. Kapitulnik, "Observation of quantum oscillations in the electrical resistivity of $SrRuO_3$," Phys. Rev. B 58, pp. (15 November 1998).

8. L. Klein, L. Antognazza, T.H. Geballe, M. R. Beasley, and A. Kapitulnik, "Possible non-Fermi liquid behavior of $CaRuO_3$," *Phys. Rev. B.* **60**, p.1448 (July 1999).

9. S. Kashiwaya, Y. Tanaka, N. Yoshida, and M.R. Beasley, "Spin current in ferromagnet/insulator/superconductor junctions," *Phys. Rev. B.* **60**, pp, 3572-3580 (August 1, 1999).

10. N.J.C. Ingle, R.H. Hammond, M.R. Beasley, and D.H.A. Blank, "The generation and detection of high flux atomic oxygen for physical vapor deposition thin film growth," *Appl. Phys. Lett.* **75**, 4162-4164 (December 1999).

11. D.C. Worledge and T.H. Geballe, "Spin-polarized tunneling in $La_{0.67}Sr_{0.33}MnO_3$," *Appl. Phys. Lett.* **76**, 900-902 (February 2000).

12. T.H. Geballe and Boris Y. Moyzhes, "On O^{16}-O^{18} Isotope Effects in Manganese Perovskites," *J. Electroceramics* **4**, 289-292 (2000).

13. D.C. Worledge and T.H. Geballe, "Maki Anaylsis of Spin-polarized Tunneling in an Oxide Ferromagnetic," *Phys. Rev. B.* **62**, 5447-451 (1 July 2000).

14. T.H. Geballe and Boris Y. Moyzhes, "Qualitative Understanding of the Highest Tc Cuprâtes," *Physica* C, **341-348**, 1821-1824 (2000).

INTERACTIONS AND TRANSITIONS

Meetings:

M. R. Beasley:

American Physical Society, Spring Meeting, Los Angeles, CA (March 1998);
"Critical Problems in Disordered Metals," Limited attendance workshop at UCLA (March 1998)
Aspen Winter Conference on Condensed Matter, invited participation, January 1999.
American Physical Society 1999 Centennial Meeting, Atlanta, GA, March 1999. Co-author of presented paper, "Spin Current in Ferromagnet/Superconductor Junctions," with S. Kashiwaya, Y. Tanaka, and N. Yoshida.
Air Force Review. Dayton, Ohio September 28-29, 1999. Attended as Principal Investigator of Air Force grant.
6th International Conference on Materials and Mechanisms of Superconductivity and High Temperature Superconductors, Houston, February 20-22, 2000. Conference organizer and session chair.
American Physical Society, Minneapolis, March 19-22, 2000.
American Superconductivity Conference, "General introduction to superconducting materials," Virginia Beach, VA. Sept. 17-22, 2000.
Topical Workshop on Interfaces and Grain Boundaries in High Temperature Superconductors, Opening Remarks and Workshop Charge, Colonial Williamsburg, VA. Sept. 25-27, 2000.

A. Kapitulnik:

American Physical Society, Spring Meeting, Los Angeles, CA (March 1998);
Aspen Center for Physics, invited participation, July 1998.

American Physical Society 1999 Centennial Meeting, Atlanta, GA, March 1999. Co-author of presented paper, "Growth of Single Crystal $SrRuO_3$ Films," with J. Reiner, A. Marshall, T. Geballe, M. Beasley.

T.H. Geballe:

American Physical Society, Spring Meeting, Los Angeles, CA (March 1998).
Limited attendance workshop at UCLA, "Critical Problems in Disordered Metals," (March 1998).
Chair of Invited Session, "Physics of Manganites Workshop," Michigan State University, East Lansing, MI. July 26-29, 1998
5th International Workshop on Oxide Electronics, December 7-8, 1998, University of Maryland, College Park, MD. T.H. Geballe and Boris Y. Moyzhes, "On O^{16}-O^{18} Isotope Effects in Manganese Perovskites."
American Physical Society 2000 March Conference, "Spin-Polarized Tunneling in Perovskite Ferromagnets," (joint presenter), March 20-24, 2000.

Consulting/Advisory Functions:

M. R. Beasley
Member, Visiting Committee to Division of Engineering and Applied Sciences, Harvard University.

Transitions:

The technology for IBAD deposition of buffer layers for use on substrates for passive rf thin film superconducting applications is being transferred to Conductus. Contact: Vladimir Matijasevic.

NEW DISCOVERIES, INVENTIONS OR PATENT DISCLOSURES:

Invention and Technology Disclosure # 98-201. "Fast Analog Correlator Based on Coupled Oscillators," M. Barahona and M.R. Beasley. (October 1998).

HONORS/AWARDS

M.R. Beasley, Fellow, American Academy of Arts and Sciences, 1991
M. R. Beasley, elected to National Academy of Sciences, 1993
A.Kapitulnik, elected to Fellowship in the American Physical Society, 1994
T.H. Geballe, American Academy of Arts and Sciences, 1972
T.H. Geballe, National Academy of Sciences, 1973